An Examination of the Evolution of the Santorini Volcanic Field Using a New Oxygen Fugacity Method

Presented in Partial Fulfillment of the Requirements for Graduation with a Bachelor of Science in Geological Sciences in the undergraduate colleges of The Ohio State University

by

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Table of Contents

Acknowledgements:

I want to dedicate this thesis to my parents who have provided me with a quality education. I would like to thank Dr. Barton for suggesting this project and providing methods and samples, answering all of my questions about source material, and the grateful review of the many parts of this thesis. Graduate student Scott Drew, and Jeff Ziga for their help with review of this thesis, abstract and poster preparations.

Abstract:

Volcanism on Santorini has occurred over the last ~600,000 years. During this interval most of the erupted lavas have evolved via processes such as fractional crystallization, mixing and assimilation, and therefore do not provide good information about their mantle source. The basalts that do occur on Santorini are parental to the evolved magmas. Studies of the basalts potentially provide information about magma genesis in the mantle beneath Santorini.

Santorini is located in the Aegean Sea above an active subduction zone. The rate of subduction is ~ 5 cm per year so that over the lifespan of the volcanic field approximately 30 km of oceanic lithosphere has been subducted into the mantle beneath the volcanic arc. As with all convergent margins, subduction of lithosphere releases fluids and produces melt in the overlying mantle wedge creating the magma responsible for volcanism. These interactions should modify the composition of the mantle over time. However, it is not clear whether the composition of the mantle wedge and hence the compositions of basaltic magmas generated from the wedge vary through time. Santorini provides a unique opportunity to address this question.

Data collected by previous workers for basalts from Santorini have been compiled and analyzed. In particular, the compositions of olivine crystals in two basalts erupted about 400,000 years apart have been used to define the pre-eruption temperatures and redox states of the host magmas. The redox state of the magma, monitored by the oxygen fugacity (fO_2), is believed to reflect that of the mantle source region. The latter should be sensitive to recycling of oxidized oceanic lithosphere via subduction. An increase in fO_2 indicates greater interaction of subducted lithosphere with the mantle. A new method, the Olivine-Melt method, developed at OSU, is being used to determine fO_2.

Graphical methods were used to examine compositional zoning in olivine and to constrain the compositions that were in equilibrium with the melt prior to eruption. This primarily involves examining variations in Mg/Fe between core and rim in individual olivine crystals.

Results indicate that the redox states of the basalts, and hence the redox states of the mantle source region have not changed over 400,000 years. Moreover, the pre-eruptive temperatures of the basalts have not changed over 400,000 years. Basalts with similar compositions have been erupted over the entire lifespan of the volcanic field, suggesting that these conclusions are valid for ~600,000 years. The results are consistent with those being obtained using other geochemical data at Ohio State. This constitutes the first study of the long-term compositions of basalts erupted at a convergent margin. The results imply either that the mantle source region has remained constant with time, or more probably that processes such as mixing homogenize the mantle-derived basalts in chambers located at the base of the crust.

Santorini Volcanic Field

Geography:

Santorini, Greece belongs to the Cycladean Archipelago located in the Aegean Sea to the north of Crete. It is part of the Aegean Arc along with Sousaki and Methana on the Greek mainland, Milos, and Nisyros, an island off the coast of Turkey. Santorini is made up of five islands: Thera, Therasia, Aspronisi, Palea Kameni, and Nea Kameni (Fig. 1). Included in the Santorini volcanic complex are the Christiana islands as well as the Kolumbo volcano. The current configuration of the islands can be attributed to the devastating Bronze-age eruption.

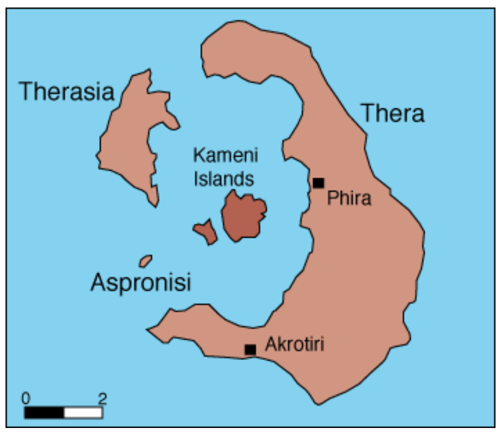


Figure1. (http://volcano.und.edu/vwdocs/volc_images/europe_west_asia/santorini.html)

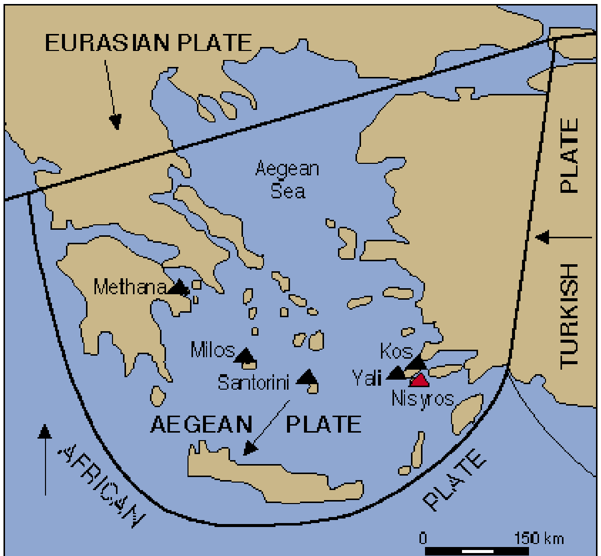
Thera is the largest remnant from the Bronze aged eruption. It is characterized by its crescent shape and includes the uplifted 200 Ma metamorphic rocks of the Cycladean Massif. The volcanoes of Akrotiri, Megalo Vouno, Mikro Profitis Elias, and Skaros consist of lava, ash, and cinder. Elsewhere, the island is blanketed with Minoan ash and pumice deposits.

Therasia is completely volcanic, is the second largest remnant of the ring island and supports a small population. Aspronisi, also known as the white island, is covered in a thick layer of the white Minoan pumice. As the island is significantly smaller, Apronisi is uninhabited in contrast to Thera and Therasia. The Kameni Islands are the products of more recent volcanic activity. Palea Kameni ("old burned") was formed in 197 BC, after the Minoan eruption (Friedrich, 2000). Due to volcanic activity and its surface structure of gaping fractures and blocky, glassy lava, Palea Kameni does not provide a hospitable environment for habitation.   Nea Kameni ("new burned") is the youngest of the five islands formed in 1707 (Friedrich, 2000). The near circular shield volcano is composed of lava, cinder, and ash. It is still warm and continues to release sulfur-rich gasses, and it's the most likely location of future volcanic activity. The caldera is a marine basin covering approximately 84.5 square kilometers, with a depth up to 400 meters below sea level (Friedrich, 2000).

Kolumbo volcano lies to the NE of Santorini only 18 meters below sea level. Its asymmetric dimensions of 4 by 8 kilometers reflect stress directions in the crust. The long axis is oriented parallel to normal faulting and extension of the crust along NE-SW tectonic lines (discussed in Tectonics). It emerged at the surface during a short period of activity between 1649 and 1650. Kolumbo's caldera has a depth of 512 meters below sea level. To the southwest lies another small group of islands included in the Santorini volcanic complex, the Christiana Islands. Today, Christiani, Askania, and Eschati are uninhabited but have ruins and other human structures dating from recent times on the largest island (Friedrich, 2000).

Tectonics:

Santorini sits on the Aegean micro-plate as part of the Hellenic Volcanic arc. This tectonic plate is moving to the southwest, while the African plate is being subducted as it travels northward towards the Eurasian plate (Fig. 2).



Volcanoes of the Hellenic arc. Arrows show direction of plate movement. Modified from Nichols (1971).

Figure 2. (http://volcano.und.edu/vwdocs/volc_images/europe_west_asia/nisyros.html)

The rate of subduction is ~ 5 cm per year (Friedrich, 2000) so that over the lifespan of the volcanic field, ~600,000 years, an estimated 30 km of oceanic lithosphere has been subducted into the mantle beneath the volcanic arc. Earthquake depths from the subducting slab define its subduction angle between 25 and 35 degrees. The Aegean arc is defined by the volcanically active centers lying above the 170 km earthquake depths (Fig. 3).

F

igure 3. From Friedrich, 2000 (after McKenzie, 1978). Aegean arc defined by earthquake depths.

Subduction provides some of the materials for magma genesis. When the subducting slab reaches a certain depth (depending on angle) it releases fluids. Consequently, this triggers melting in the mantle wedge. Fluids (eg. water) come from oceanic sediments that are not scraped off during subduction and from hydrated basaltic oceanic crust.

The varieties of volcanic rock on Santorini are created from magma mixing, fractional crystallization, and assimilation. These processes have produced a range of eruptive products from basalt to rhyodacite. As magma ascends through the 20-32 km-thick continental crust (Makris, 1978) of the Aegean plate, it melts and assimilates older crust producing the more felsic, daughter volcanics that mix with new batches of ascending magma to produce intermediate compositions. Some basalts are not subject to mixing processes during ascent and therefore have compositions that reflect the mantle source.

The relative plate movements impart a regional stress state to the Aegean micro-plate. The Aegean microplate is experiencing extension, so much that over the last 5 Ma the crustal thickness has been stretched to half of its original (McKenzie, 1978). In the Aegean region many active faults are accommodating the stresses of extension. The orientation of extension has rotated from NE-SW in the early Pleistocene to the present-day orientation, NW-SE (Druitt et al., 1999). Normal and strike-slip faulting dominate the extension of the region (Piper and Perissoratis, 2003) and in the area of Santorini; these produce a fracture system for magma to easily ascend to the surface. Shallow to intermediate depth earthquakes are focused into five main clusters beneath the volcanic centers of the Aegean arc. Earthquakes and volcanics lie on the lineated NE-SW trending geomorphic features such as grabens or troughs (Papazachos and Pangiotopoulos, 1993) (Fig. 4).

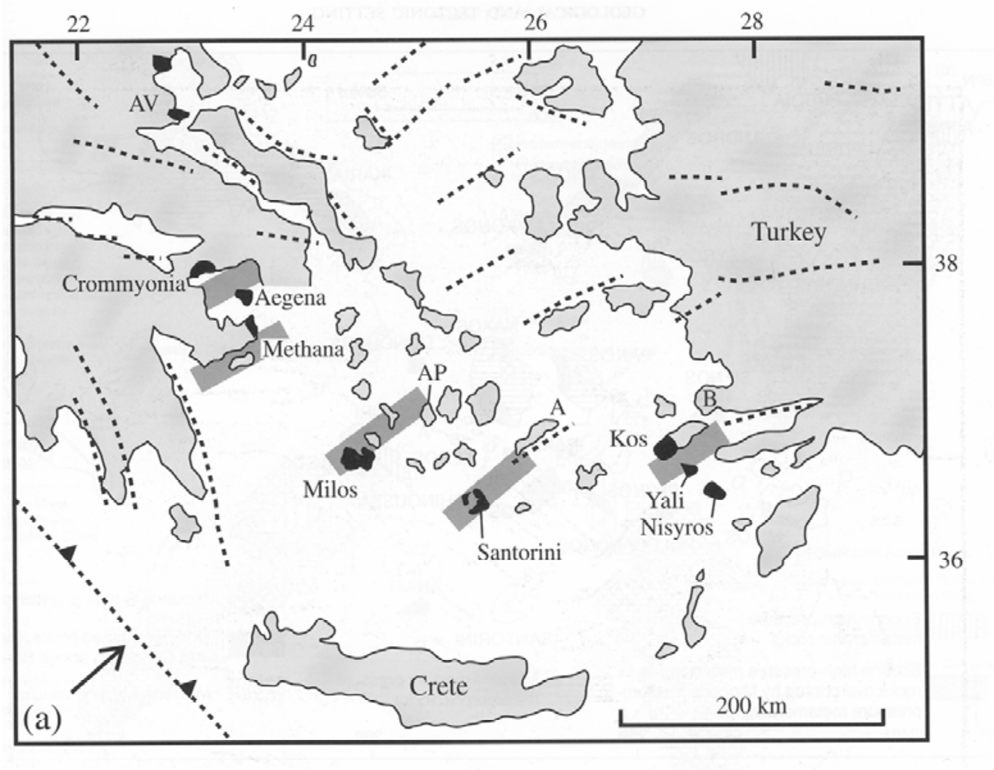


Figure 4. (Papazachos and Panagiotopoulos, 1993) Volcanic centers of the Aegean arc in black, NE-SW trending fault zone in dark grey, and active faults as dashed lines.

Tectonic lines, seen in seismic profiles, as well as in the alignment of volcanic centers on the island, follow the NE-SW trend of structural features associated with the tectonic stresses of this region. The Kameni line connects the volcanic centers of Akrotiri, Palea and Nea Kameni, and the Christiania islands to the southwest. The parallel Kolumbo line connects the Megalo Vouno and Kolumbo submarine volcanics (Fig. 5). The Kolumbo submarine volcano's long axis orientation suggests a relation between tectonic stresses and eruption.

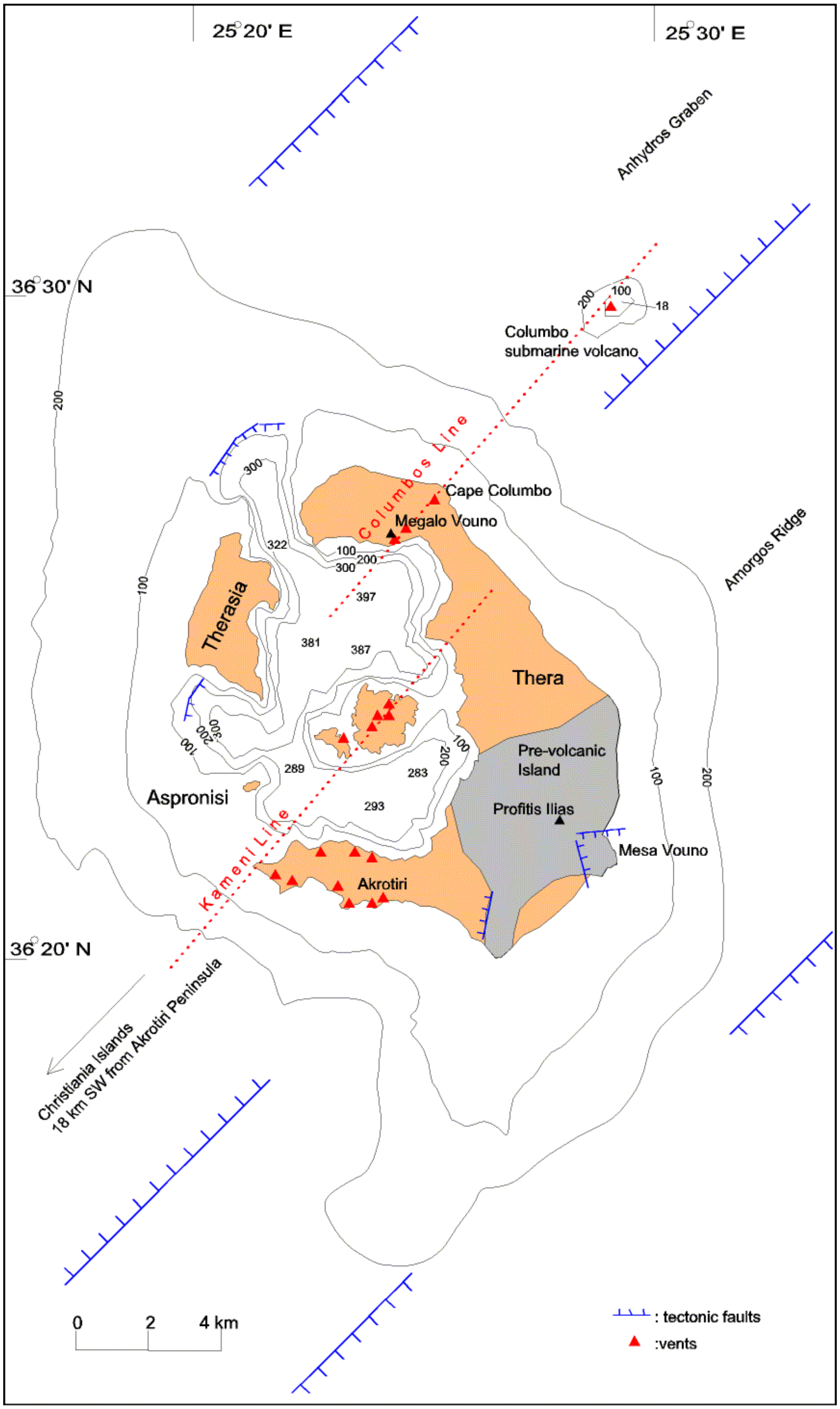


Figure 5. (http://www.decadevolcano.net/santorini/figures/santorini_caldera.gif)

Geologic History:

The oldest part of Santorini is the non-volcanic Cycladean metamorphic complex on Thera. The metamorphic complex forms the highest point of Profitis Ilias, and consists of marble, quartzite, and mica schists. The original sediments were deposited in the former Tethys Sea (Friedrich, 2000) and were uplifted during the Cenozoic as the result of tectonic stresses associated with subduction initiation. The earliest absolute age for igneous activity on Santorini is 9.5 Ma (Skarpelis et al., 1992) for a granitic intrusion into the Cycladean Massif.

The island is composed mostly of andesite to rhyodacite with a few eruptions of basalt. The numerous volcanic eruptions have produced lava flows, pumice and tuff blankets, cinder cones, and scoria eruptions. After the granitic intrusion, the first volcanics were laid down in submarine eruptions 737,000 to 563,000 years before present (Druitt et al.,1999) covering most of the metamorphic complex.

The majority of the island was created in the past 200,000 years, in which time there have been at least 12 explosive eruptions, many minor eruptions, and at least four caldera collapses (Druitt et al., 1999). The most recent eruption was a lava flow in 1950 on the central island of Nea Kameni.

One of the major eruptions or Santorini is thought to correlate with the fall of the Minoan civilization. Archeologists, historians, geographers, and geologists all suggest evidence that would link Santorini with the destruction of the city of Atlantis. A dated bronze aged settlement was discovered under the Minoan-age blanket of ash. Tradition describes Atlantis as consisting of a ring (or multiple ring) shaped island that experienced earthquakes and a flood (tsunami) that extinguished the powerful city (Friedrich, 2000).

Oxygen Fugacity

Introduction:

The fourth most abundant element in the Earth's crust is iron. Having three oxide states, it is a good indicator for oxidation state of igneous systems (Frost, 1991). Oxygen fugacity is the variable that describes the redox state of minerals and melt and is governed by the mineral assemblage or melt composition (Frost, 1991). Oxygen fugacity (fO_2) was first introduced into geologic literature by Hans Eugster in 1956, when he used oxygen buffers to control equilibria in experimental studies (Eugster, 1957). The relationship between the oxidation state of the magma source region and the oxidation state of the extruded volcanics can tell us about the igneous processes that occur during ascent (Kress and Carmichael, 1991). In order to link volcanic products and their source regions we need to know how the redox equilibrium is related to temperature, oxygen fugacity, composition and pressure (Kress and Carmichael, 1991). The oxidation state of the source region will be reflected by the oxidation state of the melt if the latter ascends as a closed system and does not substantially interact with wall rock of a different fO_2 (Kress and Carmichael, 1991).

Oxygen Fugacity plays an important role in determining the fluid phase compositions of igneous rocks (Frost, 1991), and is one of the most significant variables of basic magmas because it affects the redox state of iron in the liquid. The fO_2 of silicates is a function of Fe/Mg, Fe^{3+}/Fe^{2+}, and the Ti concentration (Frost, 1991). When fO_2 is low, iron is incorporated into silicates; when fO_2 is high, iron is incorporated into oxides (Frost, 1991). There are three reactions that clearly define this relationship, the first of which defines the elemental incorporation of oxygen to form the iron rich end member of olivine:

SiO_2	+	$2\ Fe^{0}$	+	O_2	=	Fe_2SiO4		(QIF)
quartz		iron				fayalite		

At higher fO_2, iron commonly incorporates into magnetite:

$3\ Fe_2SiO_4$	+	O_2	=	$2\ Fe_3O_4$	+	$3\ SiO_2$		(FMQ)
fayalite				magnetite		quartz		

At very high fO_2:

$4\ Fe_3O_4$	+	O_2	=	$6\ Fe_2O_3$		(MH)
magnetite				hematite		

The upper limit of oxygen fugacity for fayalite is reached in FMQ. The oxide-silicate relations above are important because these reactions are more likely to tell us about magmatic conditions than oxide reactions. The oxide-silicate assemblages have high blocking temperatures as opposed to oxides that can be reset during cooling (Frost and Lindsley, 1991). Magnesium and titanium also affect the stabilities of silicates, magnetite, and ilmenite (Frost, 1991). When Mg is incorporated into iron-silicates they are stabilized to higher fO_2. This means that even at high fO_2 both olivine and hematite can coexist if the olivine is magnesian (Frost, 1991).

The most direct measurement of fO_2 is done by sampling gases by means of inserting an electrolyte sensor directly into the cooling basaltic lava (Sato and Wright, 1966). Another method involves coexisting iron-titanium oxide compositions, or coexisting Fe-Ti oxides and silicates; unfortunately this method can only be applied to magmas that contain both ilmenite and magnetite, and in which oxide compositions have not been reset during cooling (Carmichael, 1967).

Olivine-Melt Equilibrium:

Roeder and Emslie (1970) determined the equilibrium relationships between olivine and basaltic melts as a function of temperature. Experiments at specified temperatures and oxygen fugacities were run to obtain the compositions of olivine and melt. Composition was found to be independent of temperature; the compositions of olivine crystallizing from the melt depends only on the Mg/Fe^{2+} ratio in the liquid (Roeder and Emslie, 1970). The equilibrium between Mg and Fe can be related by the distribution coefficient K_D, defined by Roeder and Emslie (1970):

$$K_D = (X^{Ol}_{FeO} / X^{L}_{FeO})(X^{L}_{MgO} / X^{Ol}_{MgO})$$

This relationship describes the partitioning of Fe^{2+} and Mg between olivine and melt.

$$Mg(Ol) + Fe^{2+}(L) \leftrightarrow Mg(L) + Fe^{2+}(Ol)$$

As a function of temperature the log of the distribution coefficient is defined by Roeder and Emslie (1970) as:

$$\text{Log } K_D = 171/T - 0.63$$

At high temperatures (1150-1300 C) the log of K_D does not vary due to the small numerator. This relation can therefore be used for magmas formed at high temperature conditions within Earth. Use of mole fractions is appropriate because deviation from ideality is too small to affect calculations. Figure 6 shows the example of the Roeder and Emslie method with $K_D = 0.3$ as a good fit for calculating olivine from liquid.

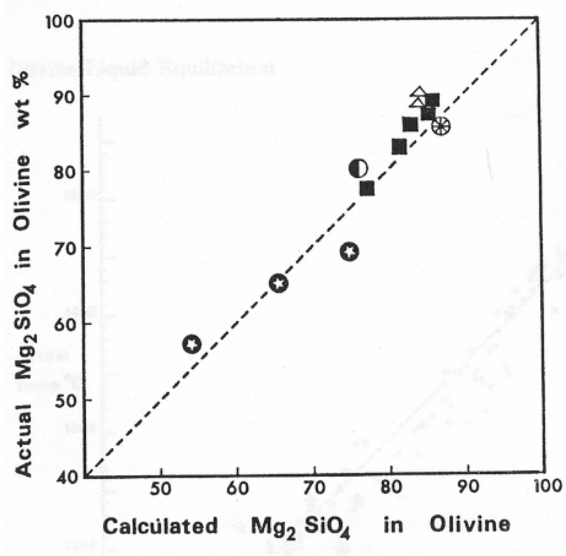


Fig. 6. Actual composition of olivine is compared to the composition which is calculated from the liquid composition using $K_D = 0.30$. The data are from the following sources: ⊛ Moore and Evans (Prehistoric Makaopuhi Lava Lake); ◐ Peck, Wright and Moore (Alae Lava Lake, Hawaii); ✪ Hakli and Wright (Makaopuhi Lava Lake); △ Green T. (Basalt Experiments 4.5 and 9.0 kb); ■ Green D. and Ringwood (Basalt experiments 4.5 and 9.0 kb)

Figure 6. (Roeder and Emslie, 1970)

The composition of olivine can therefore be used to determine the Mg/Fe ratio of the liquid from which it formed, and vice versa. The redox state of iron in the liquid is important because it will reflect the crystallization temperature and composition of the solids formed (Carmichael and Ghiorso, 1986). It is also very important to examine the temperature dependence of fO_2. Roeder and Emslies (1970) experiments show that the stability of olivine is dependent on fO_2, but that K_D is independent of fO_2.

Effect of Crystallization on Redox State

Oxygen Fugacity and the Fe^{3+}/ Fe^{2+} ratio are intimately related by crystallization. When the Fe^{3+}/Fe^{2+} ratio in the liquid is increased, due to the preferential incorporation of Fe^{2+} into crystallizing silicates (eg. olivine and pyroxene), $\log fO_2$ should increase and vice versa (Barton, personal comm.; Carmichael, 1991). This has been demonstrated by Carmichael (1967) and also for samples from the Reykjanes Ridge (Barton, personal comm. and data). As crystallization increases the Fe^{3+}/Fe^{2+} in the melt and $\log fO_2$

increases, magnetite begins to crystallize and creates a reverse relationship where Fe^{3+}/Fe^{2+} and $\log fO_2$ decrease.

In Santorini basalts we do not see a change in fO_2 during crystallization because both olivine and magnetite precipitate so that these two trends balance out (Barton, personal comm.).

<u>Olivine-melt method:</u>

The method developed by Dr. Michael Barton was used to find the oxygen fugacity of both samples SI-181 and SH-33. Figure 7 shows a summary of the method.

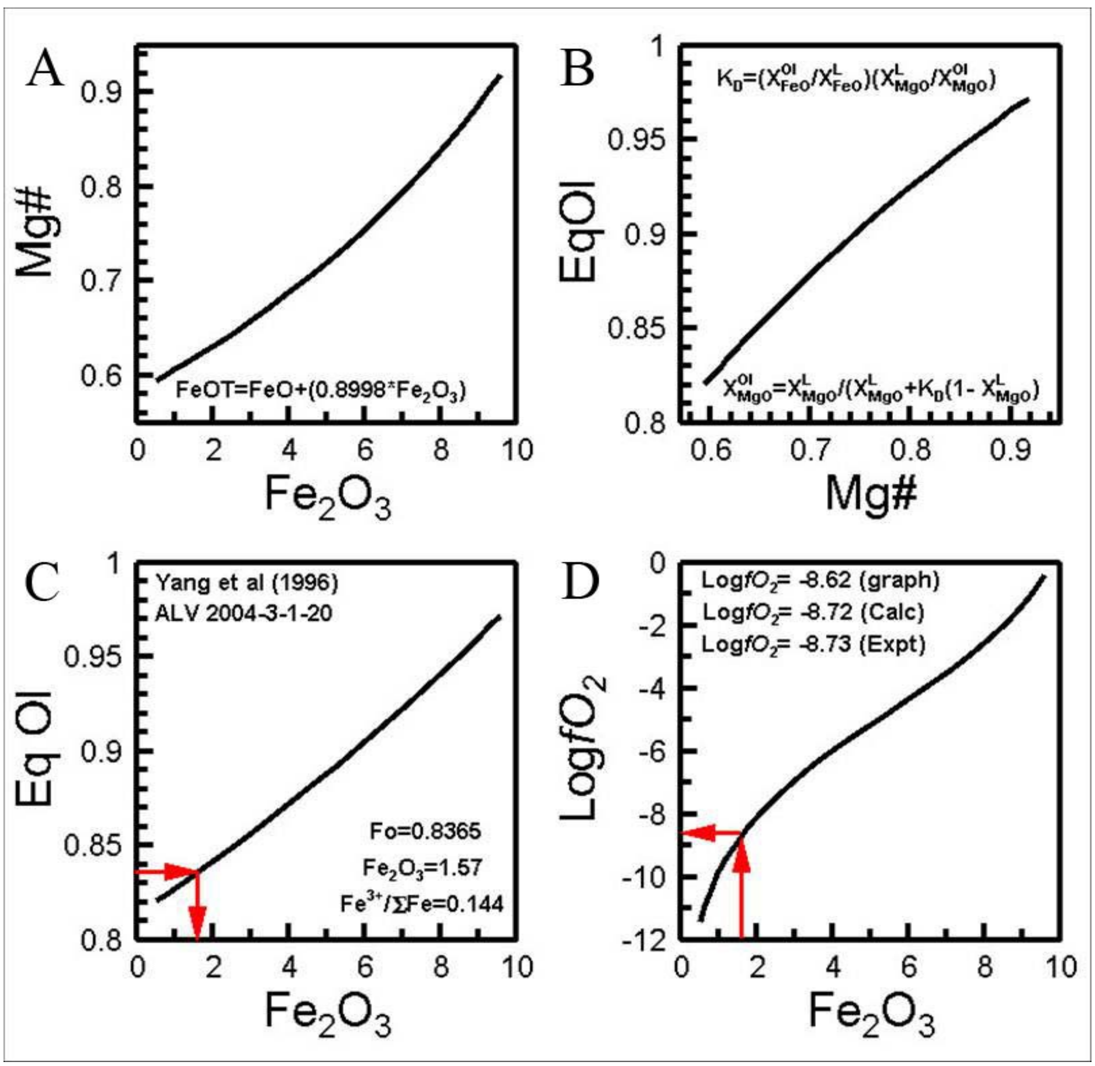


Figure 7.

Graph A shows the relationship between the Mg# and the Fe_2O_3 content of the melt where Mg# is defined as MgO/(MgO+FeO). Unfortunately data output from most analyses only report all iron as FeOT, the total iron in the sample. The method needs to distinguish ferric from ferrous iron. Graph A states that the Mg# will vary depending on the content of Fe_2O_3 in the sample.

Graph B shows that the equilibrium composition of olivine can be calculated from the Mg# and K_D. Following Miller et al. (2005), values of K_D can be predicted from melt compositions using the equations of Gee and Sack (1988).

Graphs C and D show an example of the method using a glass from Yang et al (1996). The sample is an experimental glass that was prepared at a specified oxygen fugacity, temperature, and pressure, and then analyzed. By knowing the analyzed olivine composition (Fo) we find the Fe_2O_3 in the melt. From the Fe_2O_3 content we can then solve for logfO_2 using the equation of Kress and Carmichael (1991).

$$\ln(X_{Fe2O3}/X_{FeO}) = a \ln(fO_2) + b/T + c + \Sigma_i d_i X_i$$

Where a, b, c, and d_i are experimentally determined constants. The use of this example shows the accuracy of the method. The fO_2 value is calculated to within ± 0.3 log bar units of the experimental value. Values of logfO_2 are normalized to the FMQ buffer at a specified temperature in order to compare oxygen fugacity for any basic lavas erupted at different temperatures; this is defined as ΔFMQ.

Results:

Over the lifetime of the Santorini volcanic field, approximately 600,000 years, the compositions of most of the erupted lavas do not provide good information about the mantle source region. The compositions have been modified by processes such as fractional crystallization, mixing, and assimilation within the crust. The basalts that occur on Santorini are parental to those evolved magmas, and represent the least evolved magma composition. Studies of the basalts provide most information about magma genesis in the mantle beneath Santorini, and hence about the mantle source region. These basalts have been erupted over an approximate life span of 600,000 years, over which 30 km of lithosphere has been subducted into the mantle beneath the volcanic arc. Interactions between fluids released by the subducted slab and the overlying mantle wedge should modify the composition of the mantle over time. However, it is not clear whether the composition of the mantle wedge and hence the compositions of basaltic magmas generated from the wedge vary through time. Santorini provides a unique opportunity to address this question.

The two basalts used in this study come from the southern Akrotiri peninsula and the northern Skaros lava shield. Sample SH-33 is from a massive lava-like core of a cinder cone at Cape Balos on the Akrotiri peninsula, with an age range of 522 to 340 ka (Druitt et al., 1999). Sample SI-181 is taken from the Cape Skaros lava shield with an age of 67 ± 9 ka by $^{39}Ar/^{40}Ar$ dating (Druitt et al., 1999).

The goal of these analyses was to identify olivine in equilibrium with the melt (groundmass). The olivines are zoned; the cores were crystallized at an early stage of cooling and the rims show the range of compositions that developed as the groundmass crystallized. The average forsterite contents of the olivines were examined using histogram comparisons of rim and core data from each sample. The equilibrium forsterite content was chosen by determining the most Mg (Fo) rich rim that formed as the groundmass began to crystallize (Figures 8 and 9).

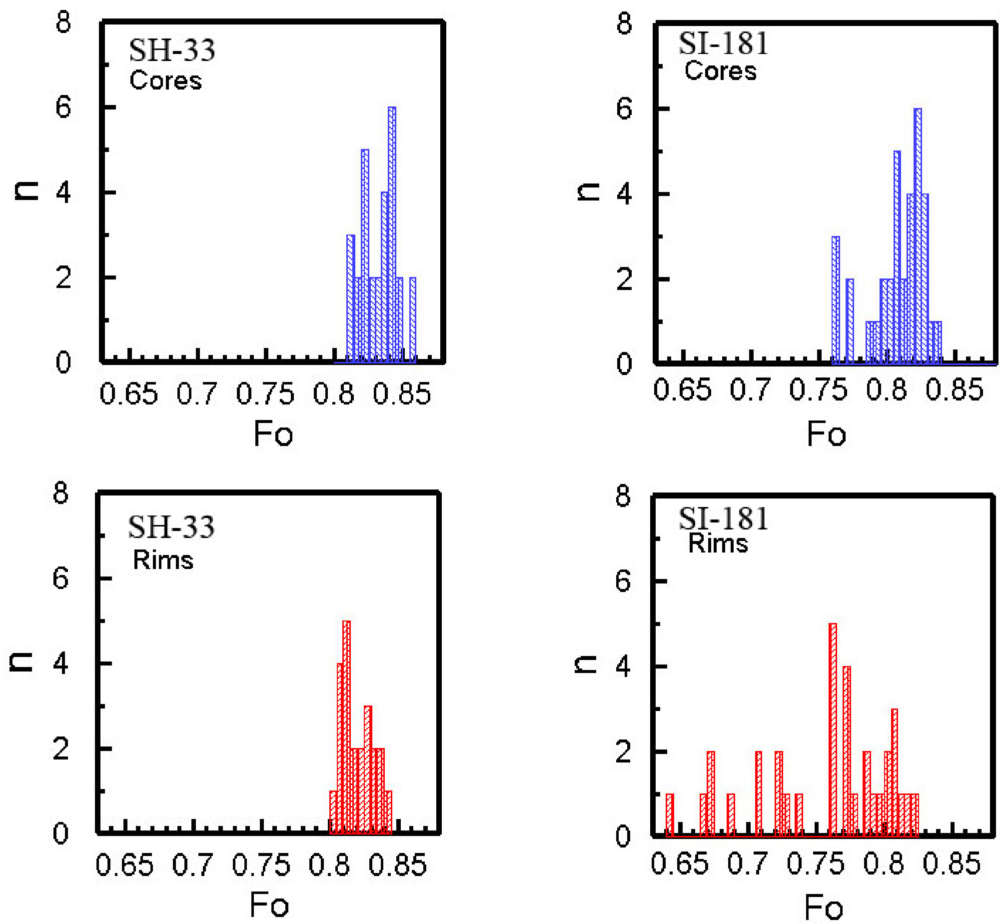


Figure 8.

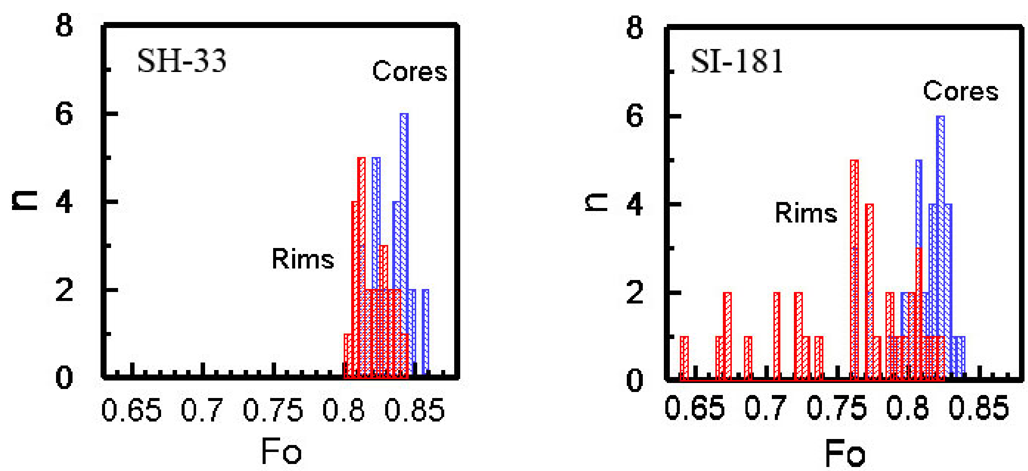


Figure 9. Olivine analysis - Microprobe data, Huismann (1985), Barton, unpublished. Groundmass - Microprobe analysis Barton, unpublished

Table 1:

Sample	Temperature	Pressure	Average Forsterite content	logfO2 (deltaFMQ-Kress)
SI-181	1074 +/- 25 C	3283 +/- 500 bars	0.8041	1.0767 +/- 0.3
SH-33	1057 +/- 25 C	3012 +/- 500 bars	0.8074	1.3566 +/- 0.3

Using the olivine-melt method, the oxygen fugacity was calculated for each sample. Temperatures and pressures were previously calculated using olivine and pyroxene compositions. Results are listed in table 1; full analyses are available upon request from Dr. Michael Barton. Thin section descriptions located in the appendix.

Discussion:

The redox states of the basalts, and hence the redox states of the mantle source region have not changed over 400,000 years. Since our logfO_2 shows an island arc signature, falling within the calk-alkaline range of oxygen fugacities, this suggests that the magma is indeed generated by interaction of the subducting slab with the mantle. Temperatures and pressures for the basalts are identical within error, suggesting that the Skaros and Akrotiri basalts were generated at the same conditions. Chemical compositions are very similar, which would support homogenization of the melt before eruption.

These results are consistent with those being obtained using other geochemical data analyzed by Scott Drew at Ohio State University. Interpretation of major and trace elements by Drew and Barton (2006) shows that that chemical composition of Santorini parental basalts (<51% SiO_2) have stayed nearly constant for ~600,000 years. Drew and Barton (2006) suggest two possible end-member explanations for this chemical constancy: 1) the primary magma has stayed near constant over hundreds of thousands of years or 2) magmas are mixing and homogenizing at the base of the crust and are periodically tapped to the surface.

The first conclusion is possible, but the results are surprising given the large amount of subducted lithosphere and rapid subduction rate. Evidence for slight isotopic differences between basaltic lavas strengthens homogenization of the magmas. These conclusions are not mutually exclusive, the range of primary magmas is small and variations can be explained by some combination of homogenization and near constant compositions.

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Appendix: Thin Section Descriptions

SI-181

Plagioclase and olivine occur as phenocrysts in a microcrystalline groundmass of olivine, plagioclase, and opaques. Plagioclase phenocrysts are subhedral to euhedral, ranging from 0.08 to 0.22mm on the long axis. Poor to moderate twinning quality and the relatively few crystals prevent compositional determination by the Carlsbad-albite method. The elongate plagioclase microphenocrysts sometimes show poor pilotaxitic texture around phenocrysts. Plagioclase phenocrysts make up a volume of ~10-15% of the sample, with another 40-45% in the groundmass. Olivine microphenocrysts are subhedral to anhedral, and range in size from 0.03 to 0.1mm. Microphenocrysts rims show a red-brown alteration in plain light. This may be iddingsite; a serpentine alteration with hematite staining; but the fibrous mineral is too small to be positively identified. Olivine microphenocrysts make up ~2-10%, with another 30-35% in the groundmass. Opaque minerals are anhedral to euhedral, less than 0.02mm, and make up 3-5%. These are possibly magnetite crystals. Clinopyroxene is rare and only present in the groundmass with sizes less than 0.1 mm

SH-33

Olivine and plagioclase occur as phenocrysts in a hyalopilitic groundmass with olivine microphenocrysts. Olivine phenocrysts make up ~10% of the sample, and are euhedral to subhedral with sizes ranging from 0.05 to 0.4mm. The rims of large olivine crystals appear to be intergrown with the plagioclase microphenocrysts. Olivine microphenocrysts make up ~20 % of the sample, and are also euhedral to subhedral with a size range of 0.03-0.15mm. Plagioclase phenocrysts make up only ~2% of the sample, are subhedral in shape, and range in size from 0.05 to 0.15mm in length. The presence of poor to moderate twinning quality and a lack of abundant crystals prevent compositional determination by the Carlsbad-albite method. The elongate plagioclase microphenocrysts show moderate pilotaxitic texture, and make up ~50% of the groundmass. Vesicles form a significant portion of the sample, contributing to 10 to 20% of the sample, with an average size of 0.05mm. Clinopyroxene is only present in the groundmass, and is rare as microphenocrysts, with sizes less than 0.1mm